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Quasi-Biennial and Long-Term Fluctuations In Total Ozone

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ABSTRACT—Quasi-biennial and long-term fluctuations in total ozone are estimated from stations with more than 4 yr of record in north polar, tropical, south temperate, and south polar latitudes and more than 8 yr of record in north temperate latitudes. The quasi-biennial total-ozone fluctuations tend to be out of phase in tropical and extratropical latitudes, with the extratropical fluctuations better organized in the Southern than Northern Hemisphere. In the Tropics, the quasi-biennial total-ozone and zonal wind oscillations are significantly in phase, but, in extratropical latitudes, the total-ozone fluctuations are alternately in phase and out of phase with the tropical zonal wind oscillation, the out-of-phase relation being the dominant one. There is the suggestion that the Mt. Agung volcanic eruption in 1963 caused a breakdown of the quasi-biennial, total-ozone oscillation in the Southern Hemisphere subtropics, with anomalously high ozone values a few months after the eruption.

We derive, for the past 15–20 yr, an increase in total ozone of 3.9 percent per decade in the Northern Hemisphere, a decrease of 1.2 percent per decade in the Southern Hemisphere, and a world-wide increase of 1.5 percent per decade. An increase in total ozone in north temperate latitudes seems quite certain, the four regional areas all showing increases varying from 1.9 percent per decade in North America to 10.7 percent per decade in the Soviet Union. There is considerable evidence that total-ozone maxima occur 1–3 yr after sunspot maxima, with the relation most pronounced in the Southern Hemisphere, but we find little evidence of a reduction in total ozone due to nuclear testing (production of nitric oxide).

1. INTRODUCTION

Evidence recently has been presented for an increase in total ozone at many stations throughout the world during the decade 1960–70 (Komhyr et al. 1971). According to a report on man's impact on climate (Wilson et al. 1971), we must establish the reality of such long-term trends so that the present and future impact of man on his environment may be accurately gauged. One of the purposes of this paper is to try to determine the extent to which total-ozone increases in some areas of the world may have been compensated by decreases in other areas of the world, and, hence, to better document the evidence for a world-wide variation in total ozone. Another purpose is to put on a firmer footing the relation between the quasi-biennial zonal wind oscillation in the lower tropical stratosphere and the quasi-biennial fluctuation in total ozone, considered earlier on the basis of shorter periods of record by Ramanathan (1963), Rangarajan (1964*a*, 1964*b*), Angell and Korshover (1964), and Shah (1967).

The annual variation in total ozone will not be considered here; instead, the interested reader is directed to the paper by Gebhart et al. (1970), which also describes an attempt to model the average world-wide distribution of total ozone and its seasonal variations. Dütsch (1969, 1970) has presented excellent summaries of most other aspects of the ozone problem and has included an extensive list of references in his 1969 paper.

2. PROCEDURES

The basic data for this analysis consist of mean monthly total-ozone amounts for individual stations throughout the world. After 1959, these data were obtained from the monthly publication, *Ozone Data for the World* (Canadian Department of Transport, 1960–1970), issued by the Meteorological Service of Canada in cooperation with the World Meteorological Organization. Ozone data for the International Geophysical Year and Cooperation were obtained from "Catalogue of IGY/IGC Meteorological Data (World Meteorological Organization 1962). Total-ozone data prior to 1957 were obtained from a variety of sources, all of which are listed in Angell and Korshover (1964).

The mean monthly total-ozone amounts are usually based on at least 10 observations during the month, but occasionally no data are obtained for the entire month. This frequently occurs in winter at stations in polar latitudes where the sun is continuously below the horizon. In this study, if one or two mean monthly total-ozone values were missing in a series, we estimated these values by linear interpolation between adjacent values, but if more than two were missing, we used the average value for that month, as determined from the total length of record. The latter results in a conservative estimate of the temporal variation in total ozone.

Our aim is to synthesize these individual station values of total ozone to obtain estimates of the total-ozone variations for climatic ozone that are as representative as possible. The values in climatic zones will in turn be synthesized to provide estimates of total-ozone variations in Northern and Southern Hemispheres and the world. Table 1 presents a listing of the total-ozone stations used in this study. Stations were selected only if their length of record exceeded 4 yr in north polar, tropical,

TABLE 1.—*Total-ozone stations used in the analysis and their period of record*

Station—latitude and longitude (deg.)	Period of record
North polar zone	
Resolute, Canada (75N, 95W)	July 1957–May 1971
Dikson Island, Russia (74N, 80E)	June 1960–Apr. 1970
Tromso, Norway (70N, 19E)	July 1935–June 1969†
Murmansk, Russia (69N, 33E)	Oct. 1961–Apr. 1970
Reykjavik, Iceland (64N,* 22W)	Jan. 1961–Dec. 1969
North temperate zone	
Soviet Union	
Leningrad (60N, 30E)	July 1957–Apr. 1970
Irkutsk (52N, 104E)	July 1960–Apr. 1970
Kiev (50N, 30E)	June 1960–Apr. 1970
Alma Ata (43N, 77E)	Nov. 1957–Apr. 1970
Europe	
Lerwick, United Kingdom (60N, 1W).	May 1952–Apr. 1970
Aarhus, Denmark (56N, 10E)	Sept. 1951–Apr. 1970
Oxford United Kingdom (52N, 1W)	Nov. 1950–Apr. 1970
Arosa, Switzerland (47N, 9E)	Jan. 1932–Apr. 1970
Rome, Italy (42N, 12E)	Apr. 1954–Apr. 1970
Cagliari, Italy (39N, 9E)	Oct. 1954–Apr. 1970
Messina, Italy (38N, 16E)	July 1954–Apr. 1970
North America	
Edmonton, Canada (54N, 114W)	July 1957–May 1971
Bismarck, United States (47N, 101W).	Jan. 1963–June 1971
Caribou, United States (47N, 68W)	Jan. 1963–June 1971
Green Bay, United States (44N, 88W).	Jan. 1963–June 1971
Toronto, Canada (43N, 79W)	Jan. 1960–May 1971
Bedford, United States (42N, 71W)	June 1963–June 1971
Boulder, United States (40N, 105W).	June 1963–June 1971
Nashville, United States (36N, 86W).	Jan. 1963–June 1971
Japan-Northern India	
Sapporo, Japan (43N, 141E)	Jan. 1958–Apr. 1970
Tateno, Japan (36N, 140E)	Jan. 1957–Apr. 1970
Kagoshima, Japan (31N, 130E)	Jan. 1958–Apr. 1970
New Delhi, India (29N, 77E)	Jan. 1955–Apr. 1970
Ahmadabad, India (24N, 73E)	Jan. 1955–Apr. 1970
Early stations	
New York, United States (41N, 74W).	Jan. 1941–Jan. 1945
Shanghai, China (31N, 121E)	Jan. 1932–Dec. 1942
Tropical zone	
Calcutta, India (23N, 88E)	Sept. 1963–Apr. 1970
Mauna Loa, Hawaii (20N, 156W)	Jan. 1961–June 1971
Kodaikanal, India (10N, 77E)	Jan. 1958–Apr. 1970
Gan, Maldive Islands (1S, 73E)	Jan. 1964–Jan. 1968
Darwin, Australia (12S, 131E)	May 1966–May 1970
Huancayo, Peru (12S, 75W)	Jan. 1964–June 1971
South temperate zone	
Pretoria, South Africa (26S, 28E)	Apr. 1964–Apr. 1970
Brisbane, Australia (27S, 153E)	Oct. 1956–Apr. 1970
Buenos Aires, Argentina (35S, 58W)	Oct. 1965–Apr. 1970
Aspendale, Australia (38S, 145E)	July 1955–Apr. 1970
Kerguelen Island (50S, 70E)	July 1959–Dec. 1968
Macquarie Island (54S, 159E)	Mar. 1963–Apr. 1970
South polar zone	
Argentine Islands, Antarctica (66S,* 64W).	June 1960–Dec. 1967
Halley Bay, Antarctica (76S, 26W)	Sept. 1956–Dec. 1967
Byrd, Antarctica (80S, 120W)	Aug. 1962–Nov. 1968
Amundsen-Scott, Antarctica (90S)	Nov. 1961–Feb. 1968

*Stations not within the given climatic zones strictly speaking, but applied to those zones to fill in data gaps.

†No data, June 1949–February 1951.

south temperate, and south polar regions, and exceeded 8 yr in the north temperate zone. Because of the considerable number of stations in the latter zone, station-groups have been formed based on geographic affinity. Even so, however, not all stations with more than 8 yr of record have been used; in regions with a multitude of stations (such as the Soviet Union), we have chosen the stations with the longest records.

The variation in total ozone in north temperate latitudes has thus been estimated by averaging together (with equal weighting) the mean ozone values determined from stations in North America, Europe, the Soviet Union, and Japan-Northern India. In other climatic zones there are not enough ozone stations to permit such a procedure; accordingly, the data from individual stations have simply been averaged to yield an estimate for the whole climatic zone.

Since total-ozone stations tend to be grouped at certain longitudes (e.g., Australia), biases may result from such a procedure and different results might be obtained by different approaches to the problem. Nevertheless, because of the current interest in total-ozone variation and the realization that spatially representative total-ozone data will only be obtained through the accumulation of years of satellite observations (Lovill 1972, Berezin and Yelanskiy 1972), we believe it worthwhile to reexamine the problem of short- and long-term variations in total ozone with the hope that, if nothing else, other scientists will be stimulated into making their own estimates of these variations.

Various problems arise in the analysis of total-ozone data, many of them having to do with instrumental uncertainties. The various difficulties experienced with the Dobson spectrophotometer have been thoroughly discussed by Komhyr et al. (1972), and electronic improvements in the Dobson instrument have been proposed by Komhyr and Grass (1972). The Dobson spectrophotometer is used almost everywhere in the world except the Soviet Union and some Eastern Block countries, where total-ozone measurements are made with a filter ozonometer. The spectrophotometer is considered much more reliable than the ozonometer (its values usually appear too low) because readings by the latter are strongly dependent upon visibility and solar elevation (Bojkov 1969). We have grouped the Soviet stations together so that their effect on the average total-ozone variation can be judged, and eliminated if desired. Oltmans (1972) has also pointed out that, during the past decade, some stations have changed the wavelength pairs used to measure total ozone, and that this change may have induced fictitious long-term trends in ozone at a few stations (New Delhi, India, for example). The ever-present problem of calibration drift of the total-ozone instruments will be considered in more detail in section 5.

Problems outside the instrumental realm include the averaging procedure to be applied when the total-ozone record is of different length at different stations. We have chosen an averaging procedure that leads to the minimum (most conservative) variation in total ozone; that is, the total ozone is held constant at its endpoint value while the

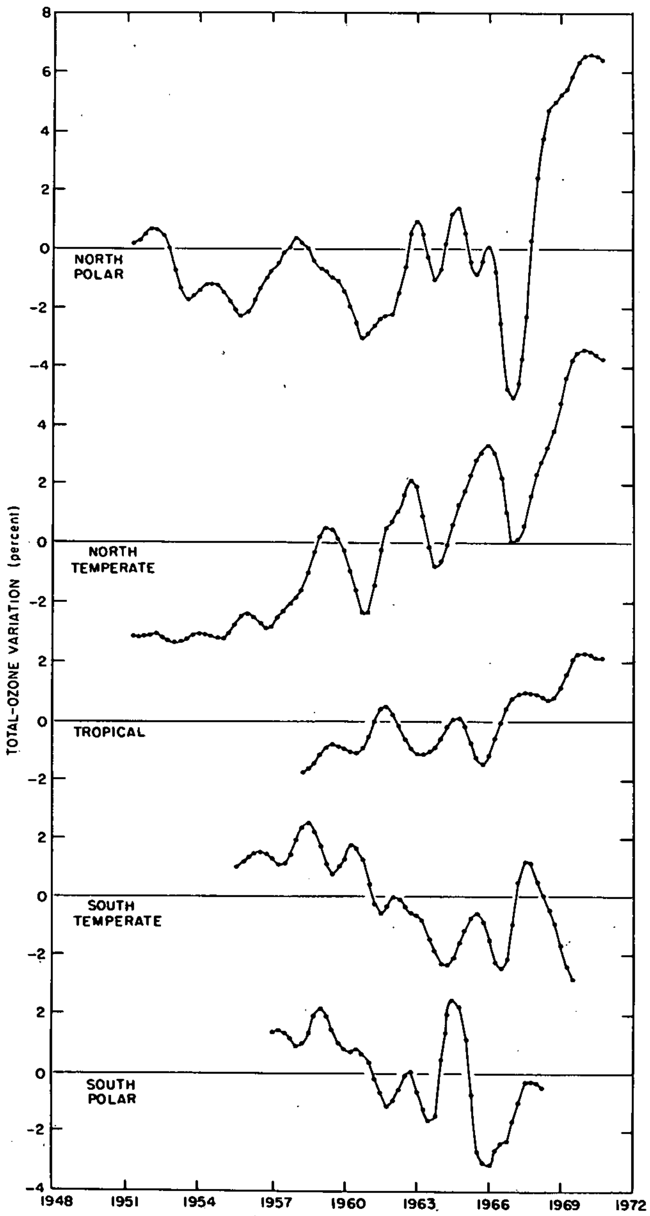

FIGURE 1.—Percentage deviation from the mean of 12-mo running average total ozone for climatic zones, as estimated from the average of individual total-ozone stations with at least 4 yr of record in north polar, tropical, south temperate, and south polar latitudes, and from groups of stations in north temperate latitudes with at least 8 yr of record. Data points are plotted at 3-mo intervals, with abscissa tick marks in June of the indicated year.

shorter records are extrapolated to the lengths of the longer records. Thus, in a sense, the variation in total ozone is weighted according to the number of stations that show a given variation.

Figure 1 presents our estimates of the 12-mo running average total ozone in the various climatic zones (expressed as percentage deviations from the mean) obtained in the above manner using the stations or station groupings listed in table 1. The 12-mo running average serves to eliminate the annual variation, which is of no interest in the present context. In this and subsequent diagrams, the data points are plotted at 3-mo intervals, and the tick marks along the abscissa represent June of the given year. Apparent in figure 1 are long-term trends in total ozone

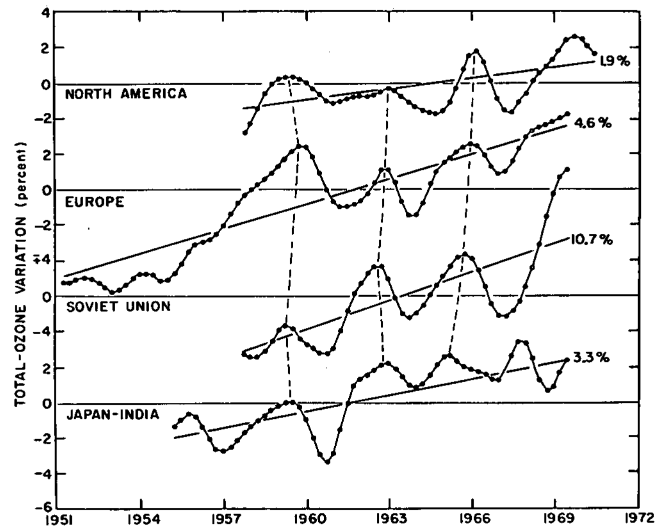

FIGURE 2.—Percentage deviation from the mean of 12-mo running average total ozone determined from north temperate latitude stations in North America, Europe, the Soviet Union, and Japan-Northern India. The dashed lines connect the respective quasi-biennial, total-ozone maxima, and the derived percentage change in total ozone per decade, obtained from the linear regression lines, is shown at right for the four geographical groupings. Note that the ordinate scale for the Soviet Union is twice that for the other groups.

as well as quasi-biennial fluctuations. In the following sections, we shall deal with these variations separately.

First, however, we wish to examine the total-ozone variations determined from the four station groups at different longitudes (and to some extent, latitudes) in the north temperate climatic zone, since this gives a "feel" for the representativeness of the total-ozone variations obtained from only a few stations in other climatic zones. The dashed line in figure 2 shows that similar quasi-biennial fluctuations in total ozone are apparent in all four groups, but there is some variation in date of total-ozone maxima and minima. We cannot be sure whether this variation in date represents a sampling problem or is real; in any case, it would appear that determination of the phase of the quasi-biennial, total ozone oscillation from stations mainly in one longitude band (e.g., Australia) should not result in phase errors of more than a few months.

Figure 2 also shows that the total ozone has tended to increase with time in all four of the above areas, with the indicated increase (based on the linear regression lines) greatest in the Soviet Union (10.7 percent per decade) and least in North America (1.9 percent per decade). This suggests that the long-term variations in total ozone obtained from the few stations in other climatic zones may at least be representative as regards the direction of trend. With all four station groups in north temperate latitudes indicating an increase in total ozone, it seems almost certain that there has indeed been an increase in total ozone in this zone during the last 15–20 yr. Note that the value of 1.9 percent per decade derived for North America is increased to 4.5 percent per decade if the regression line is determined for the period 1963–70, and this latter value is in reasonable agreement with the average value of 5.0 percent for this area for this time interval as determined by Komhyr et al. (1972).

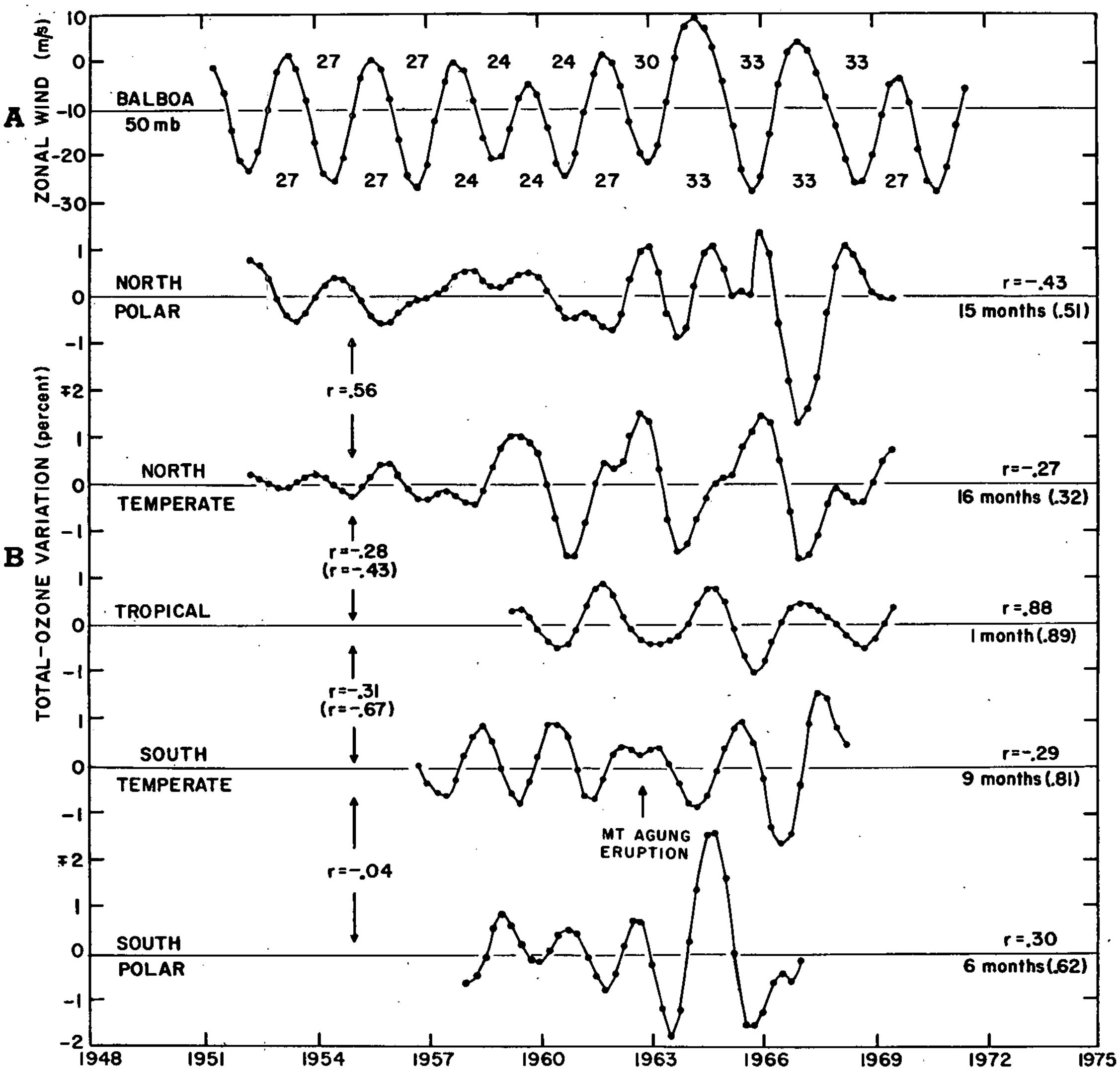

FIGURE 3.—(A) the 12-mo running average 50-mb zonal wind at Balboa (9°N) with plots of the approximate number of months between successive zonal wind maxima and minima and (B) percentage deviation of 12-mo from 30-mo running average total ozone for the climatic zones. Correlations between total-ozone variations in the different climatic zones are shown at left, with parenthetical values representing correlations of temperate values with tropical values 3 mo later. Shown at the right above the zero line are the no-lag correlations between total-ozone and Balboa zonal wind fluctuations at 50 mb; below the line is the number of months by which maximum ozone follows maximum west wind at Balboa, and the (maximum) correlation at this lag.

3. QUASI-BIENNIAL FLUCTUATIONS IN TOTAL OZONE

Before discussing our findings with respect to the quasi-biennial fluctuation in total ozone, the reader should be made aware of the recent paper on this subject by Oltmans (1972). He has examined the problem in a somewhat different way than we have; he has applied spectral and cross-spectral techniques to synoptically analyzed mean monthly maps of total-ozone amount for the Northern Hemisphere as well as to individual station data. In some cases, his results are different from those presented here. Since the data base is essentially the same, these differences point up the subtleties involved in this type of analysis. The more significant discrepancies are noted in the following discussion.

Our estimate of the quasi-biennial fluctuation in total ozone in the various climatic zones is presented in figure 3, as determined from the deviation of 12-mo from 30-mo running average total ozone. Application of a 12-mo running average to a quasi-biennial oscillation reduces the amplitude of the oscillation by about one-half, so that the ordinates in figure 3 (as well as in figs. 6–8) should be multiplied by two to obtain a realistic estimate of the magnitude of the quasi-biennial oscillation. For comparison, the 12-mo running average 50-mb zonal wind at Balboa, Canal Zone (9°N) is indicated at the top of figure 3 (the 50-mb surface was chosen because it is close to the level of maximum ozone concentration). The numbers along this latter trace, representing the approximate number of months between successive zonal wind maxima and minima, show how the tropical quasi-biennial oscillation shifted to a quasi-triennial oscillation about 1963 and then shifted back to a quasi-biennial oscillation about 1969. Total-ozone fluctuations of similar period are apparent, particularly in tropical and temperate latitudes. Bearing in mind the effect of the 12-mo smoothing, one sees in general about a 4-percent difference in total ozone at times of quasi-biennial maxima and minima. There is little evidence that this percentage changes appreciably with latitude, but since total ozone is a minimum in the Tropics, the quasi-biennial fluctuation in total-ozone amount must be greatest in polar latitudes.

The correlations between the quasi-biennial fluctuations in total ozone in the climatic zones are given left of center in figure 3. The significance of these correlations may be estimated from Fisher's Z-test (Brooks and Carruthers 1953) using the number of independent values in the series (in this case dividing the data sample by 12 because of the use of 12-mo running means). This technique gives significance levels very close to that determined by Mitchell (1966) through his concept of "effective" sample size; that is, a sample size modified according to the successive autocorrelation values of the two parameters being correlated. It is found, thereby, that correlations exceeding 0.5 are significant at the 5-percent level for record lengths of 12 yr; that is, there is less than one chance in 20 that such a correlation would be evaluated from uncorrelated populations. Figure 3 shows that:

1. Total-ozone fluctuations in tropical and temperate latitudes are out of phase, but the out-of-phase relation is not exact. For example, if temperate-latitude values are correlated with tropical values 3 mo later (values on left in parentheses), the negative correlation is considerably larger, and indeed becomes significant (2-percent level) in the Southern Hemisphere and approaches significance in the Northern Hemisphere.
2. Total-ozone fluctuations in north temperate and north polar latitudes are positively (and significantly) correlated, whereas, in the Southern Hemisphere, there is negligible correlation between these two climatic zones, suggesting a basic difference in the circulation patterns of the two hemispheres.

Indicated at the right in figure 3 (above the zero lines) are the no-lag correlations between the 50-mb zonal wind oscillation at Balboa and total-ozone fluctuations in the climatic zones. The correlation of 0.88 in the Tropics is significant at the 0.01-percent level, so there is little doubt that total ozone in the Tropics is above average in the west-wind phase of the quasi-biennial zonal wind oscillation at 50 mb and below average in the east-wind phase of this oscillation. The correlation is slightly higher (0.89) if the 50-mb zonal wind oscillation is correlated with the total-ozone oscillation 1 mo later (as indicated below the zero line at right in fig. 3). In general, we shall specify the phase relation between quasi-biennial fluctuations by evaluating the correlations at various lag intervals and then determining the time lag that yields the maximum correlation.

In north and south temperate latitudes, the total-ozone fluctuations tend to be out of phase with the tropical wind oscillations, with respective correlations of -0.27 and -0.29. Despite the similarity in these correlations, the basic relation between tropical wind and total ozone is much stronger in south temperate latitudes, as shown by the (maximum) correlation of 0.81 between the tropical zonal wind oscillation and total ozone 9 mo later, as against a (maximum) correlation of only 0.32 in north temperate latitudes 16 mo later. Thus, the similarity in no-lag correlation in the two hemispheres is the result of a weak but almost out-of-phase relation in the Northern Hemisphere in comparison with a strong but more nearly quadrature relation in the Southern Hemisphere. This tendency for the quasi-biennial oscillation in total ozone to be better organized in the Southern Hemisphere than in the Northern Hemisphere was also noted by Oltmans (1972) on the basis of cross-spectral and coherence evaluations. Application of Fisher's Z test would give a significance level of 0.1 percent for the correlation of 0.81 between tropical zonal wind oscillation and total-ozone fluctuation 9 mo later in south temperate latitudes; since we have chosen the maximum correlation from a series of correlations, however, the actual significance level is not so impressive.

Probably the most serious discrepancy between our results and those of Oltmans (1972) involves the question of whether or not the quasi-biennial variation in total ozone changes phase with latitude. Figure 3 seemingly provides good evidence that it does (at least in the Southern Hemisphere), but Oltmans claims it does not. Accordingly, it is worth examining this question in more detail. Figure 4 shows, for stations or station-groups within 50° of the Equator, the phase difference (determined from the phase lag with maximum correlation) between quasi-biennial oscillation in 50-mb zonal wind at Balboa and the quasi-biennial oscillation in total ozone. A positive phase difference signifies that the maximum in total ozone follows the west-wind maximum at 50 mb in the Tropics. The equatorial stations of Gan, Kodaikanal, and Huancayo indicate that the quasi-biennial maximum in total ozone precedes the quasi-biennial west-wind maximum by about 2 mo. It is well known that in the lower tropical stratosphere the quasi-biennial temperature maximum precedes the west-wind maximum by about 3 mo (Reed 1965). It is apparent, therefore, that very near the Equator the quasi-biennial fluctuations in total-ozone and temperature are almost in phase. Such a relation implies that vertical air motions play an important role in the temperature and total-ozone variations noted in this region; in particular, descending motions, or relatively small ascending motions, are largely responsible for the observed quasi-biennial increases in temperature and total ozone.

Figure 4 shows that, as one moves poleward in the Northern Hemisphere, the phase difference between the quasi-biennial oscillation in tropical zonal wind and total ozone appears to increase fairly uniformly, with Mauna Loa and the Japanese and (North) Indian stations indicating a phase difference about half way between that found near the Equator and that in Europe and North America. In the Southern Hemisphere, on the other hand, there appears to be more evidence for a discontinuous jump in phase, although it is admitted that the absence of total-ozone stations between 12°S and 26°S makes any such determination uncertain (in this connection, it would be useful to establish a total-ozone station near 20°S). Overall, the evidence for a phase shift with latitude of the quasi-biennial oscillation in total ozone seems almost undeniable; the phase shift appears to be progressive rather than discontinuous,

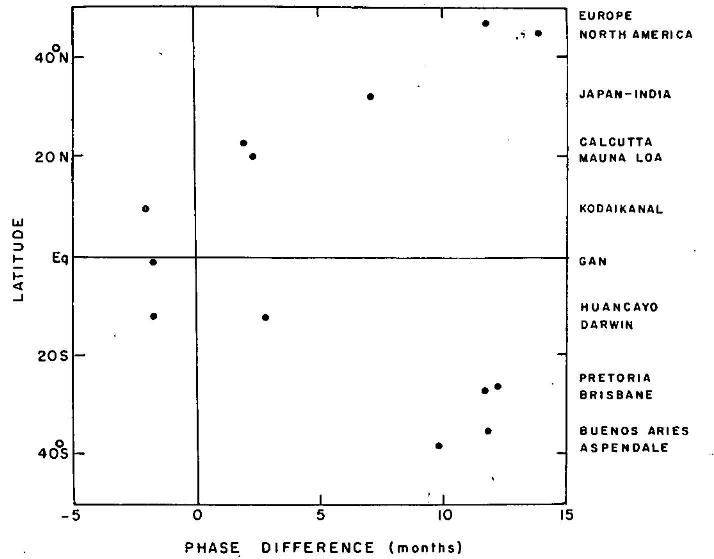

FIGURE 4.—Variation with latitude of the phase difference in months between quasi-biennial fluctuations in total ozone at individual stations or station groups and the quasi-biennial fluctuation in 50-mb zonal wind at Balboa. A positive phase difference signifies that the maximum in total ozone follows the tropical west-wind maximum.

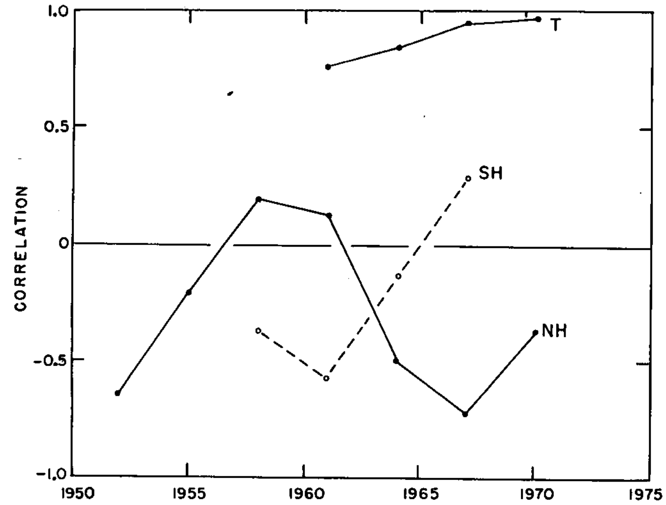

FIGURE 5.—Temporal variation in the no-lag correlation between quasi-biennial zonal wind oscillation at 50 mb at Balboa and the quasi-biennial oscillation in total ozone in the Tropics, T, and in extratropical latitudes of the Northern Hemisphere, NH, and Southern Hemisphere, SH.

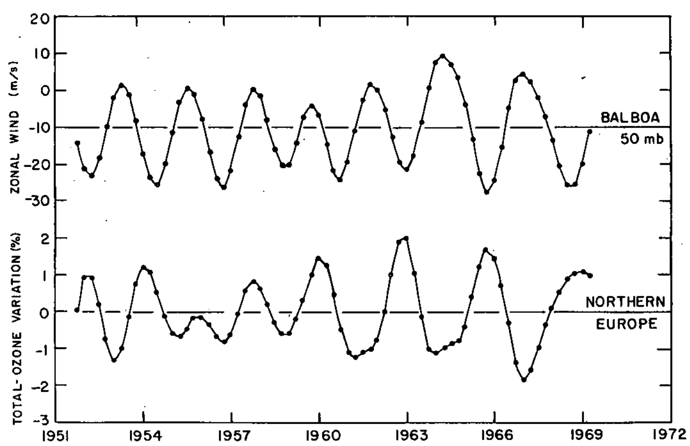

FIGURE 6.—Comparison between 12-mo running average 50-mb zonal wind at Balboa (top), and the mean percentage deviation of 12-mo from 30-mo running average total ozone at the north European stations of Lerwick, Aarhus, Oxford, and Arosa.

although this is not so clear-cut. The latter distinction is important because it would indicate whether we are dealing with a phenomenon propagating more or less uniformly poleward or with a phenomenon with a discontinuous phase jump, such as a mean meridional circulation, for example.

Figure 4 also indicates that within about 50° of the Equator the phase difference between total-ozone fluctuation and tropical zonal wind fluctuation tends to be similar in the two hemispheres (phase difference symmetric with respect to the Equator), but the correlations at the right in figure 3 show that as one progresses further poleward, an out-of-phase relation becomes much more apparent in the Northern than Southern Hemisphere. Thus, based on the lag with maximum correlation, total-ozone maximum follows tropical west-wind maximum by 15 mo in north polar latitudes but by only 6 mo in south polar latitudes. We cannot explain this marked difference in phase in the two hemispheres; as pointed out earlier, however, it would seem to imply important circulation differences in the two hemispheres. Since the phase difference between tropical wind oscillation and total-ozone oscillation in each hemisphere is slightly less in polar latitudes than in temperate latitudes, we cannot in these regions be dealing with a phenomenon propagating uniformly away from the Equator.

Careful examination of figure 3 reveals some interesting temporal variations in the phase relationship between 50-mb tropical winds and extratropical total ozone. In the Northern Hemisphere, the two oscillations tended to be out of phase after 1963 and before 1957, but in phase between 1957 and 1963. In the Southern Hemisphere, however, the oscillations tended to be in phase after 1963 but out of phase between 1957 and 1963. This result is shown quantitatively in figure 5, as obtained by a slight smoothing of the no-lag correlations between tropical wind and total-ozone oscillations at 3-yr intervals. Whereas the correlations between 50-mb tropical wind and tropical total ozone are uniformly high, the correlations between tropical wind and extratropical total ozone are oscillatory and of opposite phase in the two hemispheres. Furthermore, in the Northern Hemisphere, the above correlations are positive shortly after sunspot maximum and negative shortly after sunspot minimum, and apparently vice-versa in the Southern Hemisphere. One would certainly require more data before claiming any significance for this relationship, however. The nonsignificant coherence found by Oltmans between Ascension Island (8°S) 30-mb zonal wind, and total ozone in extratropical latitudes of the Northern Hemisphere, is probably at least partly due to the temporal variation in phase lag illustrated by figure 5.

The average of the north European stations (Lerwick, Aarhus, Oxford, and Arosa) provides a striking example of this shift in phase relationship between tropical wind and

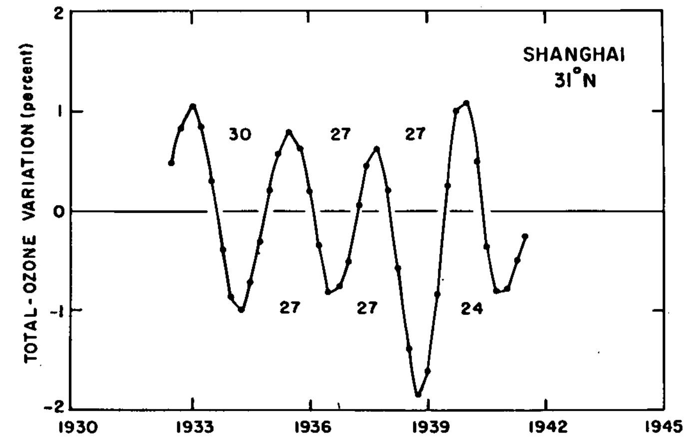

FIGURE 7.—Percentage deviation of 12-mo from 30-mo running average total ozone at Shanghai in the 1930s. The numbers indicate the approximate number of months between successive ozone maxima and minima.

total-ozone fluctuation, as shown in figure 6. Before 1954 and after 1963, the two quasi-biennial oscillations were almost exactly out of phase, whereas, between 1957 and 1960 (years of sunspot maxima), they were almost exactly in phase. There is some evidence that the exact out-of-phase relation was beginning to break down in 1969. It would be of interest for European meteorologists to examine their recent total-ozone records to see if there is any evidence that the above oscillations have again come into phase during the 1970 sunspot maximum.

Data from Shanghai, China (31°N), suggest that quasi-biennial fluctuations in total ozone occurred also in earlier years. Figure 7 illustrates that in the 1930s the period of total-ozone fluctuation at this station averaged 27 mo, but was decreasing during this time (as it did 30 yr later in the late 1960s). The (percentage) magnitude of the total-ozone variation at Shanghai is comparable to that illustrated for Northern Europe in figure 6. If the phase relation between the quasi-biennial oscillation in total ozone and tropical zonal wind were consistent, it would be possible to determine the phase of the tropical zonal wind oscillation from figure 7. However, even though recent data suggest that an out-of-phase relation is the most likely (yielding, at 50 mb in the Tropics, maximum west winds about September 1934, December 1936, March 1939, and March 1941), we cannot be sure that this is the relationship that actually existed at this time, especially since there was a sunspot maximum in 1937. There was little evidence of a quasi-biennial oscillation in total ozone at Arosa and Tromso between 1935 and 1950, implying that at middle and high latitudes of the Northern Hemisphere this oscillation may be sporadic.

4. POSSIBLE INFLUENCE OF THE MT. AGUNG ERUPTION

The largest volcanic eruption in recent years was that of Mt. Agung, Bali (8°S, 115°E), in March 1963. Newell (1970) has presented convincing evidence that a temperature increase of about 5°C occurred in the lower tropical stratosphere [e.g., at Port Hedland, Australia (20°S)]

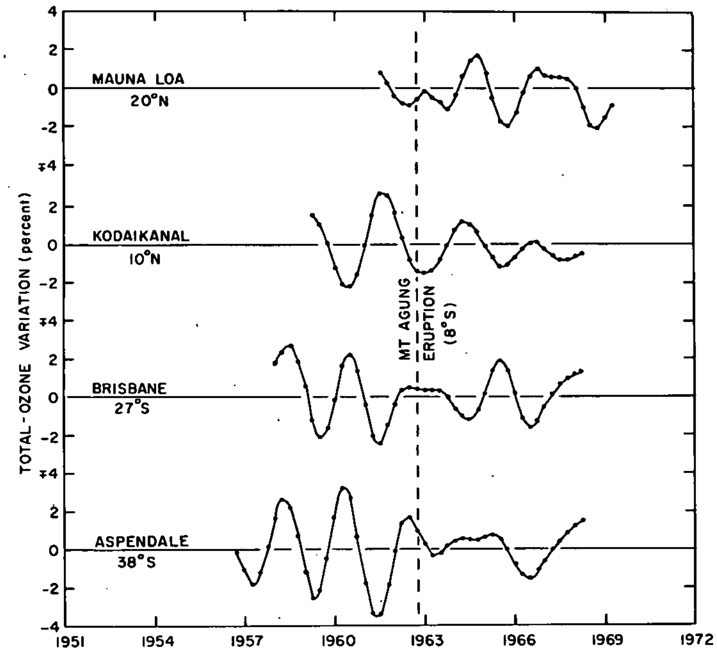

FIGURE 8.—Percentage deviation of 12-mo from 30-mo running average total ozone at the four stations closest (in latitude) to Mt. Agung and operating at the time of the eruption in March 1963.

immediately following the eruption. It is worthwhile noting the extent to which this eruption also influenced the total-ozone values. Unfortunately, in 1963 there were no total-ozone stations in operation between the latitudes of 10°N (Kodaikanal) and 28°S (Brisbane) so that, other things being equal, one would not expect any effect on total ozone to be so easily detectable.

The date of the volcanic eruption is indicated by the isolated vertical arrow in figure 3. It is of interest that in south temperate latitudes in 1963 there was a reversal in the quasi-biennial trend toward lower total-ozone values, resulting in a double-peak in total-ozone amount for the only time during the 12 yr of record. There is no obvious effect on the tropical total-ozone trace (although there was a lengthening of the period of oscillation during this time), but recall that there were no total-ozone stations in operation in south tropical latitudes in 1963. Even in the case of temperature, the eruption had little effect north of the Equator (McInturff et al. 1971).

Figure 8 presents the quasi-biennial fluctuation in total ozone at the four stations (then in operation) closest in latitude to Mt. Agung. At Kodaikanal (10°N), the amplitude of the total-ozone fluctuation has damped in recent years, but, because of the relatively short period of record, it is almost impossible to say whether this damping resulted in any way from the Mt. Agung eruption. At Brisbane, the value of total ozone remained on a relatively high plateau for a year following the eruption, part of the then widely-discussed "breakdown" of the quasi-biennial oscillation (Funk and Garnham 1962, Angell and Korshover 1964, Sparrow and Unthank 1964, Kulkarni 1966) and possibly, even likely, related to the lengthening of the tropical zonal wind oscillation at this time (fig. 3). Berson and Kulkarni (1968) suggested that the breakdown was due to the sunspot minimum occurring in 1964, whereas Newell (1972) has associated it with the Mt. Agung eruption. In any event, the oscillation resumed its normal course in 1965, and the pronounced out-of-phase relation between total-ozone fluctuation at Kodaikanal and Brisbane never was interrupted.

At Aspendale, the quasi-biennial fluctuation in total ozone broke down at least 6 mo later than at Brisbane, and the oscillation also re-formed considerably later than at Brisbane. Such a phase lag implies a poleward progression of the phenomenon that is upsetting the quasi-biennial oscillation in total ozone, whatever that phenomenon may be. Note also that prior to 1960 the total-ozone fluctuations at Aspendale clearly preceded those at Brisbane by about 3 mo (Pittock 1968), whereas since 1960 the two oscillations have been essentially in phase.

Thus, while there certainly was a breakdown in the quasi-biennial oscillation in total ozone in south temperate latitudes in 1963, it is not particularly evident from figure 8 that the breakdown was caused by the Mt. Agung eruption. In fact, careful examination of the Brisbane trace suggests that the quasi-biennial oscillation in total ozone was interrupted before the eruption; that is, the value of total ozone begins to level off in December 1962. The problem here is that the use of 12-mo running means makes it difficult to pinpoint the dates of specific occurrences.

In an effort to bypass this problem, we examined the Brisbane total-ozone data in somewhat the same way Newell examined the temperature data for this period; that is, we compared the total-ozone values for each month of 1963 with the average values for each month as determined from the 13 yr of Brisbane record. Figure 9 shows that at Brisbane, just following the time of the eruption, there was a rapid increase in total-ozone deviation from the 13-yr average, and that 3–4 mo after the eruption the total ozone was 2 percent higher than average for that time of year. This is approximately the same delay time noted by Newell in the case of temperature at Australian stations nearer the Equator. There is also agreement in the sense that both temperature and total ozone undergo a rapid increase in value followed by a gradual decline to a near-normal value. We believe that figure 9 represents strong evidence of a causal relation between the Mt. Agung eruption and Brisbane total ozone.

It is also important to consider the possibility that the Mt. Agung eruption in 1963 had a long-term effect on total ozone. However, figure 11 shows that, worldwide, a decrease or relative decrease in total ozone began about 1959, and even a 30-mo smoothing makes it difficult to relate these two events at least 3 yr apart. Nevertheless, since Pittock (1966) has shown that volcanic dust does destroy ozone by catalytic means, it is intriguing to note, from figure 12, that the total-ozone fluctuation occurs first, and in the most-pronounced fashion, in the Southern Hemisphere (the observation of an earlier and more pronounced effect in the Southern Hemisphere than in the Tropics could be due to the absence of any total-ozone stations in the southern Tropics at this time). Because of the discrepancy in dates of occurrence, we

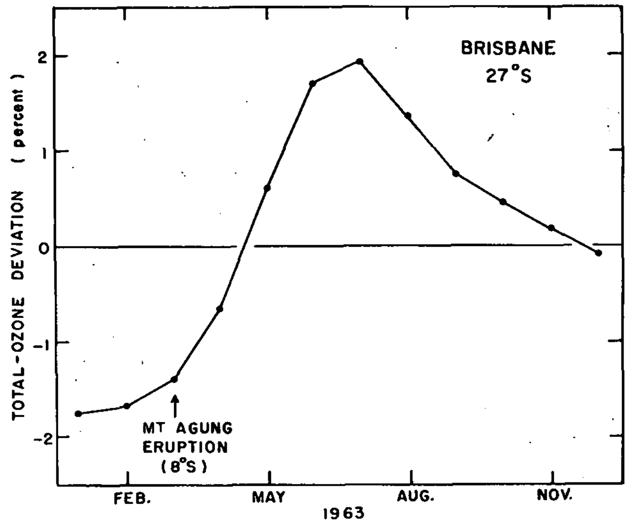

Figure 9.—Percentage deviation of the month-by-month values of total ozone in 1963 from the mean monthly values of total ozone for the 13 yr of record at Brisbane.

shall not discuss further the possibility of a relation between the Mt. Agung eruption and the long-term relative decrease in total ozone noted in figure 11, but we must admit to not being completely convinced that there isn't some relation.

If there is a relationship between the eruption of Mt. Agung and quasi-biennial, total-ozone fluctuations in nearby latitudes, how does it come about? Newell (1972) has suggested that the higher temperature and total-ozone values following the eruption could be partly explained by a "damping-off" of the Hadley circulation; for example, reduced upward motion in the tropical branch of the Hadley cell would permit atmospheric warming through radiation effects and an increase in total ozone because of a reduction in the dilution by ozone-poor tropospheric air. The difficulty with this hypothesis is that the only real evidence for ozone enrichment following the Mt. Agung eruption is at Brisbane (27°S), and perhaps Aspendale (38°S). At these latitudes, an increase in total ozone should, barring variations in the meridional eddy flux, be associated with enhanced poleward motion in the upper branch of the Hadley cell and enhanced sinking in subtropical latitudes; in other words, an *intensification* of the Hadley circulation. Actually, since one would expect some heating of the volcanic dust particles (and surrounding air) due to the net effect of solar and terrestrial radiation, an enhancement of the Hadley circulation would seem the more likely alternative. However, the suggestion of a more or less uniform increase with north latitude of the phase difference between quasi-biennial oscillation in tropical zonal wind and total-ozone fluctuation (fig. 4), as well as the slightly later breakdown of the total-ozone oscillation at Aspendale than at Brisbane (fig. 8), casts some doubt on the efficacy of the Hadley cell in this whole affair. Obviously, matters are in an unsatisfactory state; we are not absolutely sure that the eruption of Mt. Agung caused the observed breakdown in the quasi-biennial oscillation in total ozone, and if it did, we are not sure of the mechanism by which it was accomplished. This particular area of research would appear to be wide open.

5. LONG-TERM TRENDS IN TOTAL OZONE

It was pointed out in section 2 that the wavelength pairs used for total-ozone estimation changed at some stations in the 1960s, and that this may have introduced fictitious long-term trends in total ozone at a few stations. Since we shall be dealing with long-term variations in total ozone of only a few percent, there is also the possibility that instrument calibration drifts at some stations could induce fictitious long-term trends. Komhyr and Grass (1972) carried out investigations of spectrophotometer calibration drifts at United States stations in an attempt to verify the finding of Komhyr et al. (1971) of a long-term increase in total ozone east of the Rocky Mountains. They found calibration drifts at some stations and not at others, but they state that there has undoubtedly been a real increase in total ozone over the United States during the past decade. To establish the calibration drift at all the stations in the world be a tremendous, if not impossible, task. We have chosen to accept the total-ozone data as given with the expectation (hope) that any calibration drifts would be random and would cancel out in the mean. Of course, the advent of ozone measurements by satellite should eventually result in alleviation of this problem, although the present uncertainty in total-ozone measurement by satellite is about 6 percent (Mateer et al. 1971), in comparison with an uncertainty of only about 1–2 percent in the Dobson spectrophotometer if all systems are functioning properly.

Figure 10 shows the long-term trend in total ozone for the various climatic zones, as derived from 30-mo running averages of the total ozone in these zones. Thirty months was chosen because it tends to minimize the quasi-biennial fluctuations discussed in previous sections. Also included in figure 10 are the linear regression lines based on these traces, as well as the rate of change of total ozone per decade determined from the regression lines. Thus, given the total-ozone stations we have chosen to use and the manner in which we have chosen to average them, we obtain, for the last 15–20 yr, a change in total ozone of 1.8, 4.5, 2.0, −2.6, and −3.4 percent per decade in passing from north polar to south polar latitudes. The breakdown of the tropical data into regions north and south of the Equator shows that the total ozone has increased more slowly in the southern Tropics than in the northern Tropics, in general agreement with the overall tendency for an ozone increase in northern latitudes and decrease in southern latitudes.

Averaging of the four group trends in figure 2 gives a value of 5.1 percent for the average increase per decade in north temperate latitudes. The smaller value of 4.5 percent in figure 10 results from holding the endpoint values of total ozone constant in North America, the Soviet Union, and Japan-Northern India while extrapolating to the length of the European record, emphasizing again that the method we have chosen to average total-ozone records of different lengths yields a *conservative*

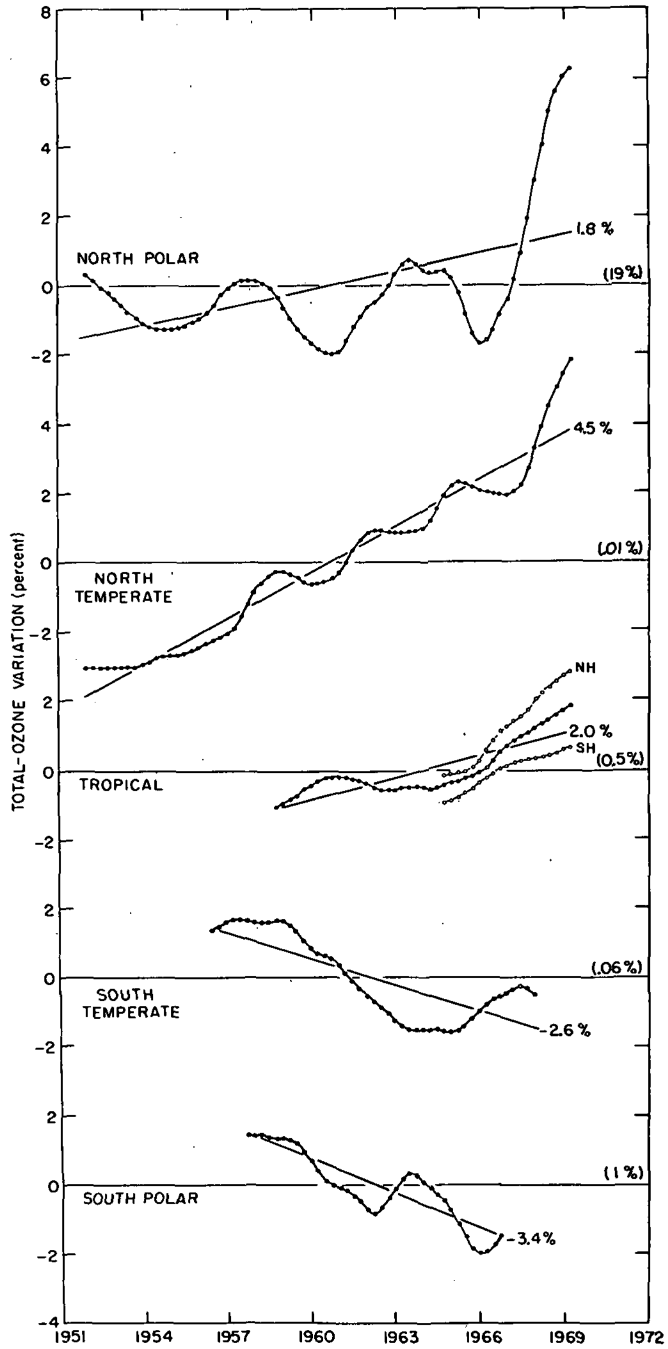

FIGURE 10.—Percentage deviation from the mean of 30-mo running average total ozone for climatic zones, and associated linear regression lines, where the numbers to the right of the regression lines represent the derived percentage change in total ozone per decade. The numbers in parentheses at far right are trend significance levels based on the Mann-Kendall rank statistic. Separate curves illustrate recent ozone variations in Northern Hemisphere, NH, and Southern Hemisphere, SH, Tropics.

estimate of the long-term trend. If one chooses to ignore the total-ozone trend at stations in the Soviet Union (because of their use of a different ozone-measuring instrument, for example), then the increase in total ozone in north temperate latitudes is reduced by about one-third; that is, from about 4.5 to 3.0 percent per decade.

The numbers in parenthesis on the right in figure 10 give the significance levels of the indicated long-term trends. The word "indicated" is used advisedly, since the significance test applies to the trace as it is plotted, and says nothing about the representativeness of that trace due either to lack of data or our manner of averaging the data. In particular, the significance determined by any technique will probably be too great because the total-ozone stations were not selected at random from a large population of stations but were selected because they were the only stations available.

With this caveat clearly in mind, the significance level was determined by means of the Mann-Kendall rank statistic (Mitchell 1966), which involves evaluating the number of subsequent values exceeding the given value at every point in the series, summing these, relating this number to a standard deviation based on the number of values in the series, and then determining the significance levels from the usual probability tables. For the individual values, we took (unsmoothed) year-average values of the total-ozone amount. This rank procedure has two primary advantages. It does not assume the trend to be linear, as do the more customary tests of significance involving the slope of the regression line, and the rank statistic is "robust"; that is, there is no assumption of a Gaussian frequency distribution.

However, the rank procedure seems to allot more significance to the trend than do somewhat more conventional techniques. For example, Pittock (1972) has presented a graph showing the significance of trends based on the use of a two-sided Student's t test. With this graph, Pittock finds that a trend we find significant at the 1-percent level is significant at only about the 5-percent level. Accordingly, our subsequent significance estimates should perhaps be scaled down somewhat.

It we apply the following terminology to the significance levels; significant if at the 5-percent level, highly significant if at the 1-percent level, and very highly significant if at the 0.1-percent level, then application of the rank technique suggests that the indicated long-term increase in total ozone in north temperate latitudes and the decrease in south temperate latitudes are very highly significant, the increase in the Tropics and the decrease in south polar latitudes are highly significant, and the increase in north polar latitudes is not significant. Note that in the north polar latitude case almost all the total-ozone increase occurred after 1966 (and much of this is associated with the Russian station on Dikson Island), in comparison with north temperate latitudes where the rate of increase apparently has been quite uniform for nearly 20 yr. Somewhat puzzling in this regard are Mastenbrook's findings, from stratospheric balloon flights near Washington, D.C. (39° N), of a 30-percent increase in water vapor at 50 mb during the years 1964–69, since it has been shown by Harrison (1970) that a 30-percent increase in water vapor would be expected to *reduce* the total ozone by several percent. However, Crutzen (1972) recently pointed out that a moderate increase in stratospheric water vapor could conceivably result in an *increase* in total ozone due to the removal of nitrogen oxides (see sec. 6) by water vapor reaction products. In the last 2 yr, the rate of increase of water vapor appears to have leveled

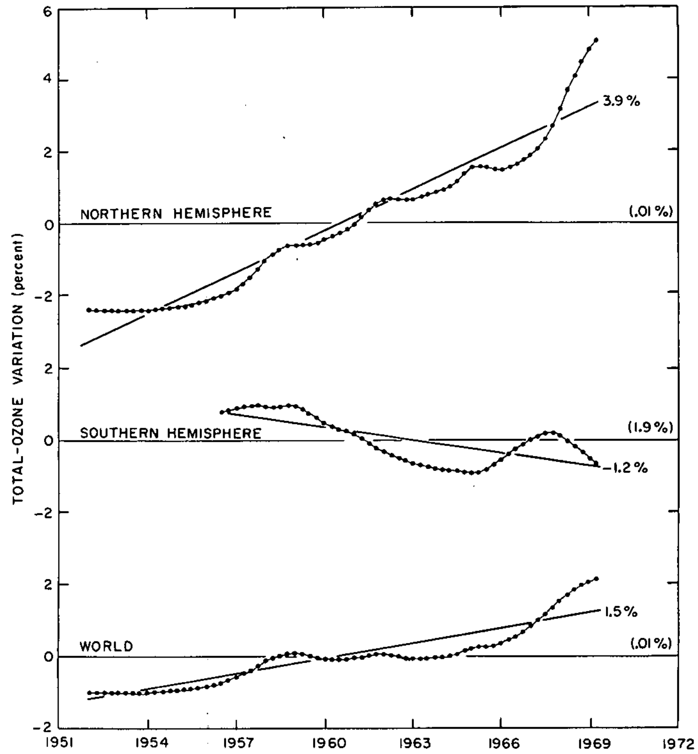

FIGURE 11.—Same as figure 10 for Northern and Southern Hemispheres and the World.

off, and it will be interesting to note the future course of the long-term trend in total ozone [the decrease in total ozone over North America beginning in 1970 (fig. 2) may simply reflect the quasi-biennial oscillation].

Figure 11 shows the long-term trend in total ozone derived for the Northern and Southern Hemispheres and for the world. The hemispheric data were obtained by weighting the trends in the climatic zones according to the surface areas of the earth they embrace; that is, Tropics 40 percent, temperate latitudes 52 percent, and polar latitudes 8 percent. The average trend for the world was then obtained by averaging the trends in the two hemispheres. The averaging of different record lengths was accomplished in the same way it was for the individual stations or station groups; that is, the shorter records were held constant at their endpoint value while being extrapolated to the lengths of the longer records. Based on total-ozone records for the past 15–20 yr, figure 11 indicates a total ozone increase of 3.9 percent per decade in the Northern Hemisphere in contrast to a decrease of 1.2 percent per decade in the Southern Hemisphere. In the world-wide average, the indicated total-ozone increase is 1.5 percent per decade. The increase in the Northern Hemisphere and the world is shown to be very highly significant (given the data and the analysis procedures used), but the decrease in the Southern Hemisphere is merely significant.

What could be the reason for an opposite trend in total ozone in the two hemispheres? The suggestion by Sleeper, Jr. (1972*b*) that the asymmetry in total-ozone trend results from the antisymmetric effects of the earth's magnetic field on charged particles emanating from the sun is an interesting one, but like most such solar-terrestrial hypotheses, it seems to be acceptable to meteorologists only as a last resort. A much more reasonable explanation, at least from a meteorological point of view, involves a temporal increase in the poleward flux of ozone in the Northern Hemisphere (either through an augmentation of mean meridional motions or eddy motions) and a temporal decrease in the poleward flux in the Southern Hemisphere. Such a strengthening and weakening of the meridional flux of ozone in the lower stratosphere might well be associated with, respectively, enhanced and reduced earthward fluxes of ozone in higher latitudes. Any increase in the northward drift of air across tropical regions associated with an interhemispheric mass exchange at ozone-rich levels would have a similar effect.

Evidence for a mean northward drift in the tropical stratosphere *has* been obtained from satellite-tracked constant-level balloon flights at 50 mb in the Tropics (Angell 1972). These balloons, launched from Ascension Island (8°S), drifted northward at a mean speed of about 0.1 m/s during the entire flight period (June 1970–March 1971). Such an interhemispheric drift would require a difference in mean vertical velocity in the two hemispheres that might possibly, through the effect of vertical motion on temperature, be reflected in long-term satellite radiance variations [see the interesting papers by Fritz (1970) and Fritz and Soules (1970) in this regard]. It is unfortunate that it is so difficult to detect weak meridional flows from conventional rawinsonde data because the possibility of a northward drift in the lower stratosphere for the past 10–20 yr (superimposed perhaps on the Hadley cell circulations) should be examined in more detail.

Given the opposition in total-ozone trend in the two hemispheres, the question next arises as to why the indicated long-term increase in total ozone in the Northern Hemisphere is not balanced by a decrease in the Southern Hemisphere; that is, why should there be a long-term increase in total ozone on a world-wide basis? Leaving aside for the moment the possibility that the data coverage in the Southern Hemisphere is simply inadequate to yield a representative estimate of the long-term trend in that hemisphere, one can conceive that the above-hypothesized mean meridional and vertical flows could bring about an increase in total ozone on a world-wide basis, although there are subtle nuances involved in such considerations. There is also the possibility that the world-wide increase in total ozone is connected with variations in solar activity, and that possibility is discussed in the next section.

6. POSSIBLE RELATION TO SUNSPOT NUMBER

It is apparent from figure 11 that the indicated world-wide increase of total ozone has not been uniform, with maxima in about 1957 and 1968 and hardly any increase between 1959 and 1965. This evidence for a nearly 11-yr periodicity in total ozone directs one's attention toward the possibility of a relationship with sunspot number. Such a possibility has been considered by many through the years. In recent years, Willett (1962) deduced, on the basis of total-ozone data prior to 1959, that there existed a world-wide maximum in total ozone 1½–2 yr before sunspot miminum. Willett's paper was severely attacked by London and Haurwitz (1963), partly on the basis of the improbability of obtaining representative world-wide averages of total ozone from the scattered data available. Later Willett and Prohaska (1965) reaffirmed their confidence in the findings of the 1962 paper but did not include any new ozone data (i.e., ozone data after 1959). In this section, we plan to reopen this particular Pandora's box. Summaries of other alleged relationships between solar activity and the terrestrial atmosphere have been presented by Lawrence (1965) and Willett (1965), and a critique of this whole problem has been prepared by Freeman and Portig (1965).

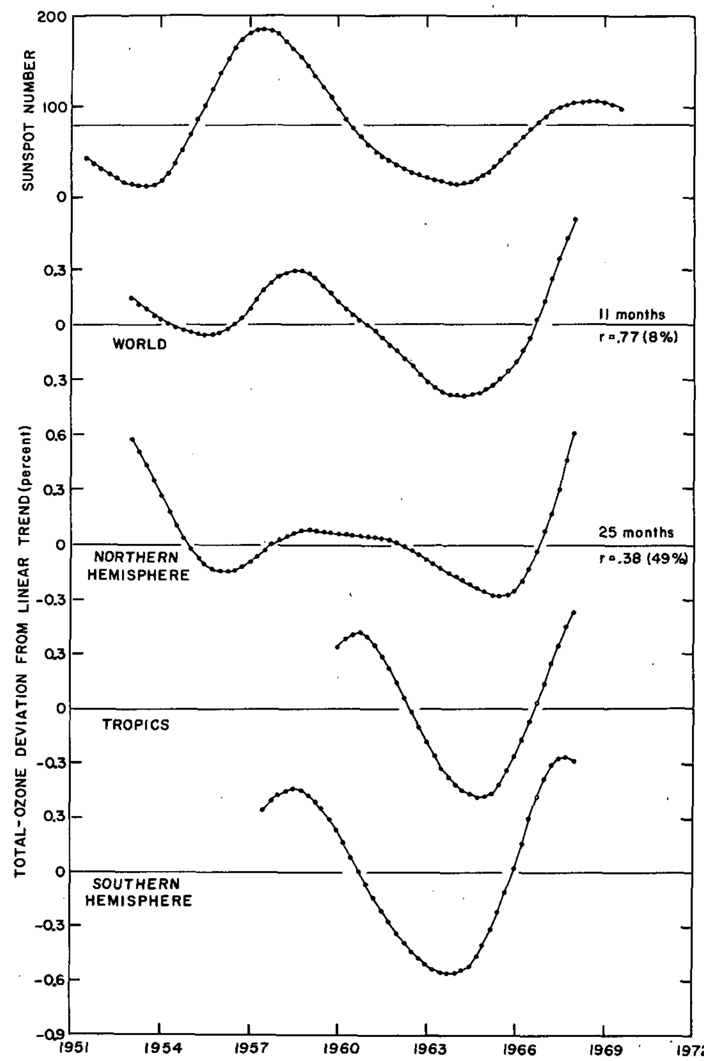

FIGURE 12.—Comparison of the temporal variation in sunspot number (top curve) with the smoothed percentage deviation of 30-mo running average total ozone from the linear trend, for the world, Northern and Southern Hemispheres, and Tropics. Indicated at right for the world and Northern Hemisphere is the average phase difference (in months) between sunspot and total-ozone fluctuations as determined from the phase lag with maximum correlation, the correlation at this lag, and the approximate significance level of this correlation (parentheses).

Figure 12 shows the comparison between the temporal variation in Zurich relative sunspot number (top curve) and the deviation, from the linear regression line, of the 30-mo running average total ozone for the world, the Northern and Southern Hemispheres, and the Tropics. On the basis of this relatively short record, there is a certain similarity between the sunspot trace and the total-ozone trace for the world, with a (maximum) correlation of 0.77 if the sunspot number is correlated with the total-ozone deviation 11 mo later. Division of the data sample by 30 (because of the use of 30-mo running averages) and application of Fisher's Z-test yields a significance level of 8 percent for the sunspot-ozone relation. As pointed out previously, however, the significance is actually not that great because we have chosen the maximum correlation in a whole series of correlations. Somewhat damaging the to concept of a meaningful relation between sunspot number and total ozone is the observation that the phase difference between the two traces has varied from nearly 3 yr in 1954 to an essentially in-phase relation after 1963. Furthermore, note that we are talking about world-wide total ozone variations of less than 1 percent, rather far into the noise level of the data.

The maximum correlation between sunspot number and total-ozone deviation is much smaller in the Northern Hemisphere (0.38), and the phase lag at which this correlation occurs is considerably greater (25 mo). As indicated in connection with the Mt. Agung eruption, comparison with the other traces in figure 12 shows that the deviation from a linear trend is greatest in the Southern Hemisphere (well over 1 percent, taking into account the effect of the 30-mo averaging) and least in the Northern Hemisphere, and that the indicated phase difference between sunspot number and total-ozone deviation is least in the Southern Hemisphere and greatest in the Northern Hemisphere. Thus, if there is a relation between sunspot number and total ozone, it appears to be showing up most clearly and most quickly in the Southern Hemisphere. Of course, there is nothing sacred about the sunspot number per se, and the relation (if any) between solar activity and total ozone may just as well result from the tendency for the charged-particle flux to be a maximum about 3 yr after sunspot maximum.

Even though we find that the relationship between sunspot number and derived world-wide variation in total ozone is not quite significant, it is a tantalizing relation and dictates the investigation of longer ozone records to determine the extent to which a similar relation holds for other sunspot cycles. Unfortunately, the two stations with the longest total-ozone record (Arosa, Switzerland, and Tromso, Norway) are in the same geographical area, and, accordingly, generalization to the rest of the hemisphere (to say nothing of the world) becomes hazardous. Figure 13 presents 30-mo running average values of total ozone at these two stations for the approximate period 1935–70 (Tromso ozone data are missing between June 1949 and February 1951, but the 30-mo smoothing has been carried through the gap), as well as the variation in sunspot number during this time. Also plotted for comparison is the derived world-wide variation in total ozone obtained from figure 11. In general, there is good agreement between the world-wide variation and the variation at

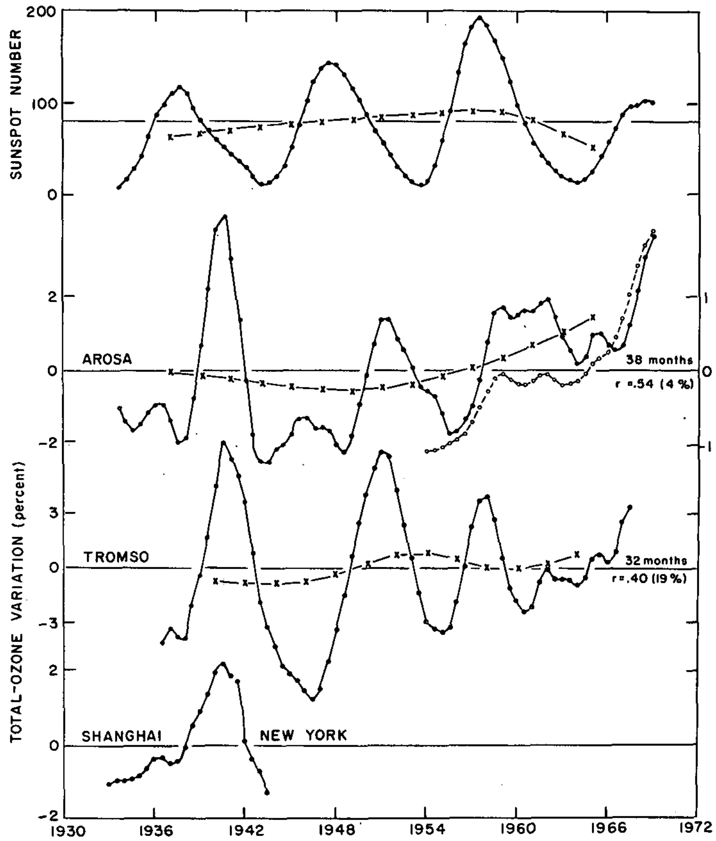

FIGURE 13.—Comparison of sunspot number (top) with 30-mo running average total ozone at Arosa (47°N), Tromso (70°N), Shanghai (31°N), and New York (41°N). The long dashed lines represent smoothed 11-yr running means; the short dashed line at right (scale at right) is the derived world-wide variation in total ozone (fig. 11). Note that the Tromso scale is twice, and the world-wide scale one-half, the Arosa scale. In this diagram, the data are plotted at either 6-mo or 2-yr intervals. Otherwise, see figure 12 legend.

Arosa, offering some hope that the Arosa total-ozone trace in earlier years provides a first approximation to the world-wide variation in total ozone.

At both Arosa and Tromso, the total ozone is a maximum about 1940 and 1951. The maxima in 1940 are the most pronounced (particularly at Arosa) and, based on limited total-ozone data for Shanghai and New York, appear to have been hemisphere-wide, but with the largest values occurring in polar latitudes (note the scale change for Tromso in fig. 13). Both the 1940 and 1951 maxima in total ozone occurred slightly after the time the sunspot number passed through its mean value on a downward trend; that is, there was slightly more than a one-quarter cycle (actually about 3-yr) phase lag between sunspot number and total ozone, as if basically the total ozone continued to increase as long as the sunspot number was above average. This is the same relation found by Dütsch (1969) for ozone variations above 30 km at Arosa. Willett (1962) obtained a phase lag ½–1 year greater than we did, a result perhaps to be anticipated based on Willett's use of only the very early total-ozone data and the evidence from figure 12 that sunspot number and total ozone have been coming more in phase in recent years.

Note, however, that despite their proximity, Tromso and Arosa are not in agreement with respect to the total-ozone maximum about 1960, the total ozone at Arosa remaining at a relatively high level almost 3 yr longer than at Tromso, emphasizing again the hazards of generalization from individual station data. The result is that, while the nearly one-quarter cycle phase lag between sunspot number and total ozone has continued at Arosa during the past decade, a more nearly in-phase relation has developed at Tromso. As a consequence, the phase lag with maximum correlation (0.54) at Arosa turns out to be 38 mo (in comparison with 33 mo for an 11-yr sunspot period with a quarter-cycle lag), while at Tromso the phase lag is reduced to 32 mo and a correlation (at this lag) of 0.40. With the usual reservation regarding the significance of the maximum correlation obtained from a whole series of correlations, the correlation of 0.54 at Arosa is significant at the 4-percent level, but the correlation at Tromso is not significant.

There is the suggestion of an inverse relation between the amplitudes of the total-ozone oscillation and the 11-yr sunspot oscillation. Thus, in 1940, 1951, and 1960, the total ozone at Arosa and Tromso averaged 6.0, 3.9, and 2.5 percent, respectively, above the 11-yr running average, whereas the sunspot numbers averaged 52, 63, and 90 above the running average. The unusually rapid increase in total ozone on a world-wide basis that began in 1966 (fig. 11) and the reduced sunspot maximum in 1970 suggest a continuation of the inverse relationship between sunspot amplitude and total-ozone amplitude, and, on the basis of the small phase lag in recent years between sunspot maxima and total-ozone maxima (fig. 12), we anticipate a pronounced maximum in the 11-yr total-ozone cycle in 1971 or 1972. However, while Paetzold et al. (1972) also show a nearly in-phase relation between sunspot number and the amount of ozone between heights of 20 and 30 km at European stations during the past 10–15 yr, they find (based on two solar cycles) a *direct* relation between the amplitude of the ozone and 11-yr sunspot oscillations, just the opposite of the result found here.

Since there is some evidence that total ozone varies with the 11-yr sunspot cycle, it is also natural to consider the possibility that longer term variations in total ozone (e.g., the increase during at least the last 20 yr) also reflect variations in sunspot number. There is an 80- to 100-yr cycle in sunspot number called the Gleissberg cycle (Sleeper 1972), which involves about four successive sunspot maxima of relatively small magnitude followed by about four successive sunspot maxima of relatively large magnitude. As anticipated by Willett in his 1965 paper, the sunspot maximum in 1970 was much smaller than preceding maxima due to the operation of this cycle (there is usually a slow increase in magnitude of the sunspot maxima followed by a sudden "break" to a low level). However, any relation between the Gleissberg cycle and total ozone seems unlikely because the smoothed sunspot number began to decrease already in the early 1960s, and a 10-yr lag between smoothed sunspot maximum and smoothed total-ozone maximum seems excessive. Furthermore, association of total ozone with the Gleissberg cycle implies that the total ozone has been increasing for nearly 40 yr, certainly not in agreement with the long-term trend in total ozone at Arosa (fig. 13). Thus, the indicated long-term increase in total ozone does not appear to be related to a variation in solar activity, even though there is some evidence that the shorter term (11 yr) variations are so-related.

There are at least two basic difficulties with the preceding inference of a relation between the 11-yr cycles of sunspot number and total ozone. The first involves the disagreement between the derived phase lag of 11 mo between sunspot number and world-wide total ozone during the past 15–20 yr (fig. 12) in comparison with the derived phase lag of about 35 mo between sunspot number and Arosa and Tromso total ozone during the past 35 yr (fig. 13). It is apparent from figure 12 that the considerable discrepancy results both from a tendency for a more in-phase relation during recent years as well as for the phase lag to be larger in the Northern Hemisphere than in the Southern Hemisphere. Since there is a possibility, however remote, that these temporal and spatial variations are real, the indicated discrepancy in phase lag may not be completely destructive to the concept of a true relationship.

The second difficulty is that, while it has been shown experimentally that there exists a relation between increased sunspot activity and enhanced solar radiation in the extreme ultraviolet (Tousey 1963, Anderson 1965), and even in the ultraviolet (Heath 1971), it has also been shown theoretically that the enhanced ultraviolet radiation is unlikely to bring about directly an increase in total ozone (Lindzen 1965, London 1969). However, a series of important papers by Crutzen (1970, 1971) and Nicolet (1972) indicate that nitrogen oxide may have considerable influence on the ozone content of the atmosphere. In addition, Johnston (1972) has shown recently that the ozone in the atmosphere is about half that to be expected from the ultraviolet flux and that nitrogen oxides are likely responsible for this "ozone deficit." Since the amount of nitrogen oxide could be affected both by ultraviolet and charged particle fluxes, a subtle association between solar activity and total ozone is not completely unreasonable. In any event, while coincidence certainly may be involved, the indicated relationship between sunspot number and total ozone in figures 12 and 13 is not easily dismissed, and we believe further investigation along this line is warranted as long as it is clearly understood that the implied response of total-ozone variation to sunspot number (or the subsequent maximum in charged-particle flux) is at most a few percent, with the response apparently being largest in polar or near-polar latitudes and probably larger in the Southern Hemisphere than in the Northern Hemisphere.

7. POSSIBLE RELATION TO NUCLEAR EXPLOSIONS

Johnston (1971) has proposed that the oxides of nitrogen in supersonic transport (SST) exhaust could, by catalytic means, reduce the ozone shield by a factor of about two.

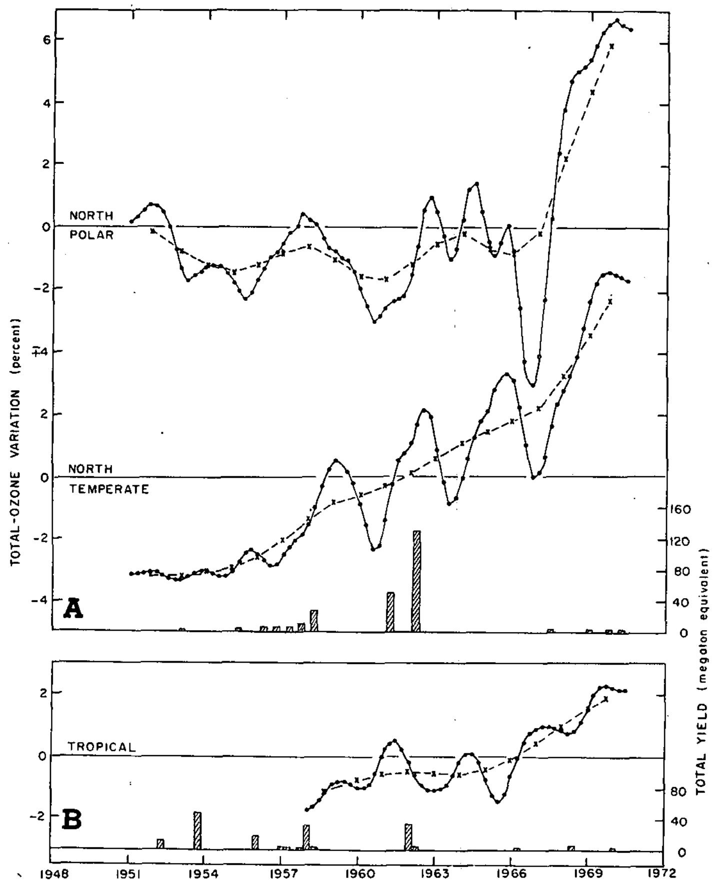

FIGURE 14.—Percentage deviation from the mean of 12-mo and 3-yr (dashed line) running average total ozone in comparison with dates and magnitudes of nuclear explosions, (A) in north polar and temperate latitudes and (B) in the Tropics.

Foley and Ruderman (1972) pointed out that, during years of intense nuclear testing, significant amounts of nitric oxide (at least 10^{34} molecules) were probably injected into the stratosphere due to intense heating of the air by the nuclear explosions. However, they found no evidence of a reduction in ozone following the periods of most intense nuclear testing and concluded that, "the ozone content of the atmosphere is probably not particularly sensitive to the presence of nitric oxide." This seemingly contradicts Johnston's association of the "ozone deficit" with nitrogen oxides, and in rebuttal Johnston et al. (1972) recently have provided observational evidence *for* a reduction in total ozone following large-scale nuclear tests. In this section, we examine this question using our estimates of total-ozone variation in climatic zones.

Figure 14 shows the relation between date and magnitude (megaton equivalent) of the nuclear explosions and the derived 12-mo and 3-yr running averages of total ozone in north polar and temperate latitudes and in the Tropics. Because of uncertainties concerning the point of origin of some of the earlier explosions in the Soviet Union, the explosions in north temperate and polar latitudes have been combined. This should not be too damaging to the analysis since one would expect that, within a few months, the mixing between the two climatic zones would be quite complete. A 3-yr rather than 30-mo running average has been used here to more effectively eliminate the quasi-triennial, total-ozone oscillations of north temperate latitudes. We indicate both 12-mo and 3-yr smoothings because of the uncertainty concerning the time scale of any nuclear effect on total ozone; that is, the response of ozone to nitric oxide injection should be almost instantaneous in the immediate area, but, because of the relatively slow mixing, any effect on the total-ozone average for a climatic zone might not be apparent for weeks, and for a hemisphere for months. An added complication is that most of the large Soviet tests occurred in autumn in polar latitudes, and the subsequent lack of sunlight might postpone for several months the photochemical effects associated with the destruction of ozone.

The solid lines in figure 14 show that the very large Soviet explosions at Novaya Zemlya (75°N) in the autumns of 1961 and 1962 took place during a period of total-ozone increase in both north polar and north temperate latitudes. At least in north temperate latitudes, that increase is part of the pronounced quasi-triennial oscillation in total ozone in existence between 1957 and 1968. These large explosions had no noticeable affect on this quasi-triennial oscillation in north temperate latitudes, but, in north polar latitudes, the total ozone trace is sufficiently complex that it is difficult to say whether there is or is not an influence. There is also no obvious effect on the longer term trend (dashed line), the rate of increase of total ozone in north temperate latitudes being relatively small from 1959 to 1967. In north polar latitudes, however, there actually was an increase in total ozone between about 1960 and 1963.

Looking at the smaller explosions in north extratropical latitudes between 1956 and 1958, one could make a case for an effect on the long-term trend. There was a decrease in total ozone in the polar latitudes after 1958 and, as mentioned above, the rate of increase of total ozone in temperate latitudes decreased after 1959. However, with the absence of an obvious effect from the later and larger explosions, one would presume that the relation in 1958 is pure coincidence.

In addition, the quasi-biennial oscillation in total ozone in the Tropics is seemingly unaffected by the explosions in 1958 and 1962. From the point of view of the long-term trend, however, the invariance in total ozone in this region between about 1961 and 1964 could possibly be related to the explosions at Christmas Island (2°N) in the summer of 1962, although the trace certainly appears to have leveled off before the explosions. The question also arises as to why there is an apparent long-term increase in total ozone following the equally large explosions in the Marshall Islands (11°N) in the summer of 1958. In this regard, note the possible confusion between cause and effect resulting from the fact that the Mt. Agung eruption (sec. 4) occurred only 9 mo after the Christmas Island test in 1962, and that both these events occurred about 3 yr after sunspot maximum. As noted earlier, this necessarily leads to uncertainty as to whether the leveling-off in the total ozone trend between about 1960

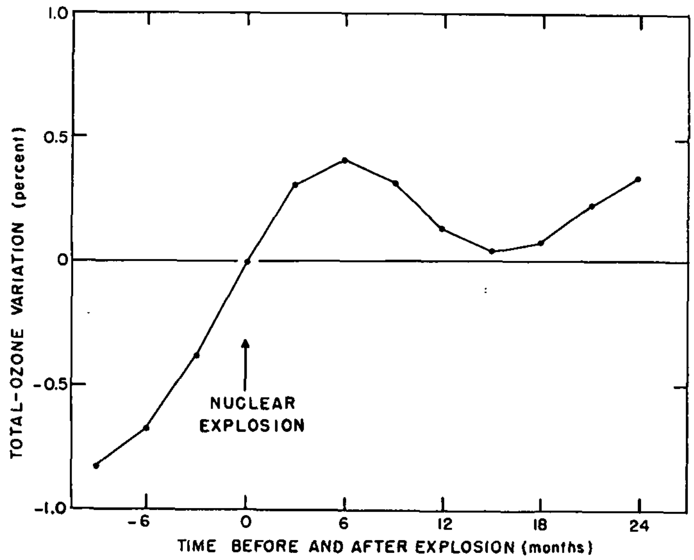

FIGURE 15.—Average percentage deviation from the mean of total ozone before and after nuclear explosions, as obtained by normalizing the ozone data to the date of the explosion and weighting the ozone variation according to the magnitude of the explosion.

and 1965 is the result of solar variations, nuclear testing, the eruption of Mt. Agung, or even some other phenomenon.

In an attempt to make the above discussion more objective, we have normalized, to the dates of the nuclear explosions, the total-ozone variations before and after the explosions, and then weighted these ozone variations according to the explosion magnitude. Figure 15 shows the derived average variation in total ozone 9 mo before and 24 mo after the normalized explosion date, where the polar and temperate-latitude total-ozone traces have been averaged for explosions in extratropical latitudes. As suggested previously, figure 15 shows that, in the mean, the nuclear explosions happened to take place at a time when total ozone was increasing rapidly, and, for the first 3 mo following the explosion, there is no evidence of a reduction in this rate of increase. After 3 mo, however, there is a reduction in the rate of increase; after 6 mo the total ozone begins to decrease, on the average. For the entire 2 yr following the nuclear explosions, it never falls below its value at the normalized time of explosion. Thus, it would appear most likely that the small decrease in total ozone occurring 6–15 mo after the normalized explosion date merely reflects the naturally occurring quasi-biennial variation in total ozone superimposed on a general upward trend in total ozone. It is our opinion that there is little evidence from either figures 14 or 15 to indicate an influence on total-ozone amount through the production of nitric oxide by nuclear explosions; this may, therefore, cast some doubt on the significance of nitrogen oxides as regards the total-ozone balance of the stratosphere in general.

We have now to explain why Johnston et al. (1972), using more or less the same total-ozone data, should find evidence for a decrease in total ozone following nuclear testing. They place considerable emphasis on the observation that, for the 27 stations with at least 30 mo of total-ozone data during 1960–62, and at least 30 mo of data between 1963 and 1970, 20 exhibited a decrease in total ozone during 1960–62 and 22 exhibited an increase in total ozone between 1963 and 1970. They then infer that the decrease in ozone in the early 1960s and the increase in the late 1960s are due, respectively, to the bomb testing in 1952–62 and the absence of appreciable testing after 1962. The increase in total ozone after 1963 is part of the long-term increase in total ozone noted in the Northern Hemisphere (all but three of the 27 stations are in the Northern Hemisphere) during the last 20 yr (fig. 11). Of the 20 stations showing a decrease in total ozone during 1960–62, nine are European stations; it is apparent from figure 2 that 1960–62 happened to be a period of considerable decrease in total ozone (3.2 percent in 2 yr on the average) in Europe due to the quasi-biennial (actually quasi-triennial) oscillation in ozone. The quasi-triennial oscillation is also responsible for a somewhat smaller decrease in total ozone during this period at North American stations, but, at stations in the Soviet Union and Japan-Northern India, this oscillation occurs earlier, causing the average oscillation derived for north temperate latitudes (fig. 14) to show little variation in total ozone between 1960 and 1962. This points up the peril of averaging the available station data rather than averaging by geographical areas; Johnston et al. (1972) have obtained a somewhat unrepresentative result because so many of the available total-ozone stations were in Europe (the inclusion of 3 stations in the Southern Hemisphere where there is an overall downward trend in ozone also helps confuse their results). In summary, we see no reason to relate the indicated downward trend in total ozone in 1960–62 to anything but the quasi-triennial oscillation in total ozone, which has persisted very strongly in north temperate latitudes for at least 10 yr.

The other important point made by Johnston et al. (1972) is that the greatest decrease in total ozone in 1960–62 occurred north of 50°N, where the largest nuclear explosions took place. This result would be anticipated, however, from the observation that the quasi-biennial oscillation in total ozone has been quite pronounced at the Northern European stations (fig. 6) and that 1960–62 happened to be a period with a sharp decrease in total ozone due to this oscillation.

In many ways, the world-wide invariance in total ozone between 1959 and 1965 (fig. 11) would appear to represent more impressive evidence for some sort of nuclear (nitric oxide) influence on total ozone. This lack of change superimposed on an overall upward trend in total ozone could be associated (with considerable time lag) with nuclear testing in 1952–62. However, as shown in figures 11 and 12, this particular effect is much more pronounced in the Southern Hemisphere than in the Northern Hemisphere and is, therefore, unlikely to be associated with nuclear testing. Furthermore, Johnston et al. (1972) state that ". . . if artificial nitric oxide from nuclear bombs is correlated with Sr_{90} and C_{14}, one expects ozone to decrease between 1960 and early 1963 and to increase from 1963 to the late 1960's." From figures 11 and 12, however, we find no evidence for an ozone increase before the middle of 1964 at the earliest, and the above authors agree, stating, ". . . this conspicuous minimum of ozone in the middle 1960's is a recurring pattern." It is this world-wide oscillation in total ozone that we, of course, have related to the sunspot cycle and were earlier tempted to relate to the eruption of Mt. Agung.

8. CONCLUSIONS

The following are the significant points with respect to the quasi-biennial fluctuations in total ozone during the past 15–20 yr:

1. There is a significant correlation of −0.67 between total-ozone fluctuations in south temperate latitudes and the total-ozone fluctuations 3 mo later in tropical latitudes. The associated correlation of −0.43 in north temperate latitudes is not quite significant at the 5-percent level. In general, there is an out-of-phase relation between total-ozone fluctuations in tropical and extratropical latitudes.
2. There is a significant and positive correlation between quasi-biennial fluctuations in total ozone in north temperate and polar latitudes, but a negligible correlation between these fluctuations in south temperate and polar latitudes, implying a basic hemispheric difference in circulation.
3. There is a highly significant correlation of 0.88 between tropical fluctuations in total ozone and the fluctuations in 50-mb zonal wind at Balboa; that is, the total ozone in the Tropics is above average in the west-wind phase of the quasi-biennial oscillation and, it turns out, a maximum almost exactly at the time of quasi-biennial temperature maximum.
4. In extratropical latitudes, the total-ozone fluctuations are alternately in phase and out of phase with the tropical zonal wind oscillation, the phase relationships being opposite in the two hemispheres. The out-of-phase relation appears stronger and more persistent. This variation in phase has a near 11-yr period (hence might possibly be related to the sunspot cycle) and is beautifully expressed by the North European total-ozone stations.
5. The breakdown in the quasi-biennial, total-ozone oscillation in south temperate latitudes occurred approximately at the time of the Mt. Agung (8°S) eruption in 1963. A causal connection is suggested by the later date of the breakdown at Aspendale (38°S) than at Brisbane (27°S), as well as by the anomalously high values of total ozone at Brisbane a few months after the eruption.
6. Total-ozone data for Shanghai in the 1930s show that the quasi-biennial oscillation existed also at this time, with periods of oscillation decreasing from 30 mo in 1934 to 24 mo in 1940.

The following are the significant points with respect to the derived long-term trends in total ozone:

1. In north polar, north temperate, tropical, south temperate, and south polar latitudes during the last 15–20 yr, we derive total-ozone changes per decade of 1.8, 4.5, 2.0, −2.6, and −3.4 percent, respectively, yielding total-ozone changes in Northern Hemisphere, Southern Hemisphere, and world of 3.9, −1.2, and 1.5 percent per decade, respectively.
2. A total-ozone increase in north temperate latitudes seems assured, with increases per decade of 1.9, 4.6, 10.7, and 3.3 percent in North America, Europe, the Soviet Union, and Japan-Northern India, respectively.
3. During the last 15–20 yr, there has been a correlation of 0.77 between sunspot number and the world-wide variation in total ozone 11 mo later, the latter determined as the deviation from the 1.5 percent per decade linear trend. The relation appears strongest, and the phase lag smallest, in the Southern Hemisphere.
4. Total-ozone records at Arosa (47°N) and Tromso (70°N) between 1935 and 1970 show that a maximum in total ozone usually follows a maximum in the 11-yr sunspot number by about one-quarter cycle (33 mo). The discrepancy with respect to the phase lag on a world-wide basis (point 3), while certainly damaging to the concept of a real relation, may be at least partly due to temporal and spatial variations in the phase lag.
5. The magnitude of the 11-yr maximum in total ozone appears to be inversely related to sunspot number. Since the sunspot maximum in 1970 was very weak due to operation of the 80- to 100-yr Gleissberg cycle, one would hypothesize an unusually pronounced total-ozone maximum in 1971 or 1972 with the above association.
6. We find little evidence of a reduction in total ozone due to the production of nitric oxide by nuclear tests, either on a case-by-case basis or by normalizing the total-ozone data to the dates of the explosions and weighting the total-ozone variations according to the magnitude of the explosions.

Finally, it is hoped that this discussion has directed the reader's attention to the complex nature of the total-ozone variation, both in space and time. Because of uncertainty concerning the raison d'être of much of the variation, it is extremely difficult at this time to evaluate accurately man-made influences on ozone amount. Consequently, when considering the possible effect of the supersonic transport on stratospheric ozone, for example, we must be very careful that any changes noted reflect the human influence and would not have occurred naturally. For conscientious scientists, this may be the most difficult determination of all.

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